

Chromatic control in coextruded layered polymer microlenses

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Abstract: We describe the formation, characterization and theoretical understanding of microlenses comprised of alternating polystyrene and polymethylmethacrylate layers produced by multilayer coextrusion. These lenses are fabricated by photolithography, using a grayscale mask followed by plasma etching, so that the refractive index alternation of the bilayer stack appears across the radius of the microlens. The alternating quarter-wave thick layers form a one-dimensional photonic crystal whose dispersion augments the material dispersion, allowing one to sculpt the chromatic dispersion of the lens by adjusting the layered structure. Using Huygen's principle, we model our experimental measurements of the focal length of these lenses across the reflection band of the multilayer polymer film from which the microlens is fashioned. For a 56 μm diameter multilayered lens of focal length 300 μm , we measured a $\sim 25\%$ variation in the focal length across a shallow, 50 nm-wide reflection band.

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1. Introduction

Microlenses and microlens arrays play increasingly significant roles in miniaturizing optical systems for beam shaping and homogenization [1], interconnections, imaging, and displays. These arrays involve patterning of micron and submicron structures on functionalized and structured materials, with a rich history of success based on the invention of a wide variety

of patterning techniques such as holographic photolithography [2], e-beam lithography [3], hot embossing/injection molding [4, 5], and imprinting [6] or microcontact printing [7] with or without using solvents. A review of top-down and bottom-up patterning techniques applied to polymers can be found in [8]. Popular methods of fabricating microlenses include the use of thermal reflow [9, 10], micro droplets [11], and imprint molding [6]. In addition, the use of grayscale photolithography has received recent attention due to several of its advantages. For example, the use of grayscale avoids the misalignment issues that arise from the use of multiple masks. Grayscale photolithography is less susceptible to distortion [12] than soft- and nano-lithography imprint techniques. Further, as described below, the use of grayscale lithography enables axial cutting and thus axial gradient index microlenses, allowing new applications not possible with a radial gradient index profile obtained by imprint molding. Use of axially-cut multilayers may enable better control over the step size in a diffractive lens so that light diffracted from increasing ring diameters etched into the lens travels the same fixed distance to the focal point. (See [13].)

Patterning using a grayscale mask followed by plasma etching to transfer the pattern to the substrate has been used for diffractive and refractive microoptics for beam shaping, spectral separation, phase modulation, lensing and other applications [14, 15]. The mask used in grayscale photolithography is itself made using high energy beam sensitive (HEBS) glass, with the pattern being traced by electron-beam lithography. The exposure to high energy electron beams causes the reduction of silver ions in the glass. Areas of the mask exposed to a higher dose from the electron beam will have higher concentrations of reduced silver ions resulting in increased optical density (OD) there. By adjusting the grayscale levels in the pattern, the curvature can be controlled so that in a single exposure, a plano-convex photoresist shape can be obtained, circumventing exposure to heat used in a thermal reflow or a hot embossing process, which may otherwise be detrimental to some polymers. By this technique, “axial” gradient index microlenses can be fabricated whereby the index variation is along the lens axis and also appears on the lens sag. This is in contrast to “radial” gradient index where the index variation is radial outward from the center of the lens and can be fabricated by imprint molding. In the axial gradient microlenses, the index variation along the lens sag can be employed to precisely tailor the angle of refraction in order to provide superior corrections of different types of aberrations [16, 17].

At the same time, layered polymer stacks formed by co-extrusion [18] are being developed for a wide variety of optics applications, such as filters [19], sensors [20], switches and optical limiters [21], data storage media [22], and lasers [23]. These multilayer materials are attractive for their ready functionalization, ease of processing and amenability to large-area, low-cost fabrication [24–26]. Most of these applications involve quarter-wave thick alternating layers of different refractive index forming a one-dimensional photonic crystal with a characteristic reflection band that depends on the layer thicknesses, refractive index, and number of layers [27]. Thus, the optical dispersion of the lens is augmented by the dispersion characteristic of the layered photonic crystal medium. This provides the ability, through careful choice of materials and structure, to sculpt the chromatic response of the microlens elements.

We report here on a grayscale mask-based etching technique applied to a multilayer polymer stack to demonstrate the technical feasibility of making microlenses with large, designed chromatic dispersion. Beyond the process of fabricating these lenses, we characterize the focusing and spectral dispersion properties of an axially-terminated microlens array etched from the multilayer polymer film possessing a shallow reflection band in the visible. A straightforward physical optics model qualitatively captures the salient features of these unique optical elements and may be useful for designing more complex layered lenses.

2. Materials and methods

The transparent constituent polymers used consisted of bilayers of polystyrene (PS) with an average refractive index of 1.6 and polymethylmethacrylate (PMMA) with an average refractive index of 1.5 made from a melt-process co-extrusion technique described in detail in [27]. The lens material had 32 bilayers, 64 individual layers in total, each layer about 80 nm thick, with an average layer-to-layer thickness variation of 22%. The total thickness of the polymer material was about 10 μm . About 2 cm \times 2 cm piece of the polymer sample was mounted on a clean silicon substrate for support and the top protective polyethylene layer was removed. A drop of thick photoresist (AZ P4620) was applied on the sample which was then spun at 2,500 rpm for 30 seconds on a Brewer Cee 200 spin coater, and baked at 90°C for 1 minute on a hot plate. Note that this baking temperature is below the T_g for PS (95°C), PMMA (\sim 100°C) and AZ P4620 (125°C). The sample was then exposed to UV light through the grayscale mask (described below) for 30 seconds using a Karl Suss MJB3 mask aligner and the sample was continuously swirled for about 2 minutes in a diluted AZ 400K developer solution, followed by rinsing in de-ionized (DI) water.

The grayscale glass mask [28] was designed such that, for a single microlens in the array, the optical density (OD) decreased radially outward through several gray levels from a value of 1.2 at the center to 0 at the edge so that the intensity of the UV light passing through the mask is varied accordingly. Figure 1(a) shows the microscope image of the grayscale mask in which one can observe the variation in the shade across the circular cross-section, representing the variation of the OD in the circular structure. Each gray level was defined by a 1.0 μm wide ring so that for a 60 μm diameter microlens, there were 30 gray levels whose OD values decreased from 1.2 at the center by 0.04, as illustrated in the optical density profile shown in Fig. 1(b). As further detailed by Canyon, Inc [28], the linearly varied OD values across the 1.0 μm -wide rings as shown in Fig. 1(c) can produce a smooth convex surface profile due to inherent proximity effects in the lithographic method and depending on the exposure parameters. Thus, the resulting photoresist pattern is convex in shape following developing in the AZ 400K developer solution. The single exposure using the grayscale mask produces a convex shape without melting the AZ P4620 photoresist which would have required a temperature that could have ruined the multilayer stacks in the polymer material.

Successful transfer of the plano-convex resist to the underlying multilayered polymer requires careful optimization of the etch parameters to remove both the photoresist and the polymer at about an equal rate. In our process, the optimum condition consisted of using a mixture of O_2 and CF_4 flowing at 2.0 and 3.0 standard cubic centimeters per minute at standard temperature and pressure; a total pressure of 100 mTorr and a radio frequency power of 20 watts. At these settings, an etch duration of 25 minutes succeeded in removing all the photoresist, thereby transferring the resist's post-exposure plano-convex shape into the multilayered polymer.

The lenses are made in square arrays that have a center-to-center separation distance of about 180 μm . Arrays of microlenses of four different sizes with lens diameters of nominally 80, 60, 40 and 20 μm were fabricated. The experimental data described here are from the 60 μm and 80 μm diameter microlenses.

Figure 1(c) is the atomic force microscope image (AFM Agilent Model 5500, friction in contact mode), which shows that the resulting lens is cut from the multilayered polymer with a ring pattern that is determined by the (quasi-uniform) layer thicknesses and the lens shape in relief. The ring pattern was similarly observed in SEM images (variable pressure scanning electron microscope imaging using the JEOL, model JSM-IT300LV, not shown) of these microlens arrays. Figure 1(d) is the 2-dimensional AFM scan across a microlens showing that this particular example was etched to a depth of about 3.5 microns.

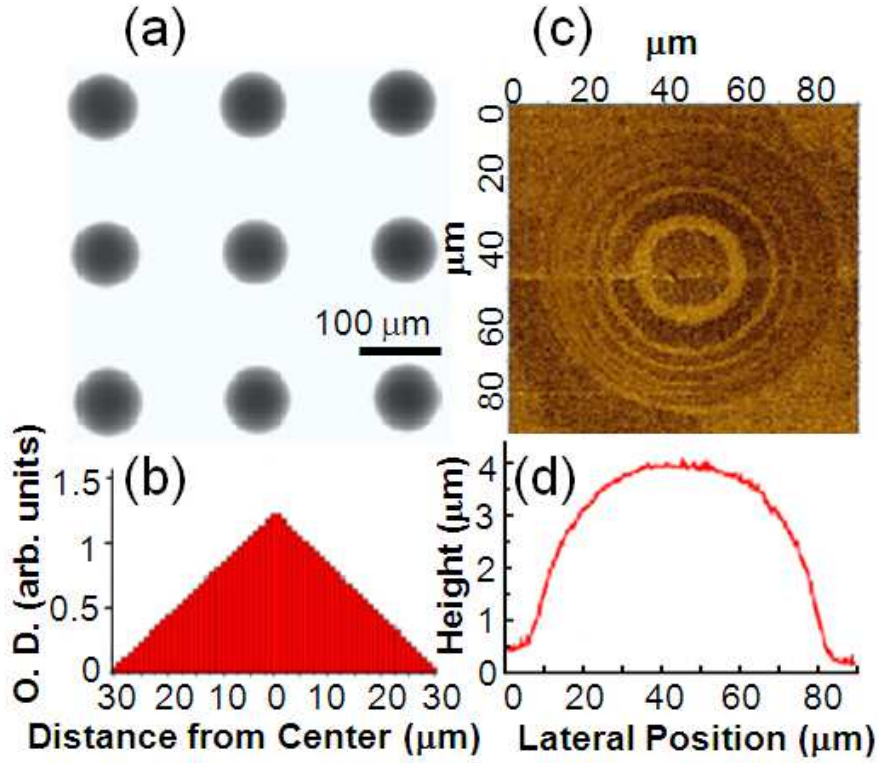


Fig. 1. (a) Microscope image of grayscale mask (top left) with (b) radial optical density (O.D) profile (bottom left). (c) Polymer multilayer microlens AFM image viewed in contact mode reveals the salient layering of the different polymers (top right) and (d) a typical polymer multilayer microlens AFM cross-section (bottom right).

3. Optical characterization

Our experimental set up for measuring the focal dispersion of a microlens is shown in Fig. 2. The particular microlens in the array to be tested is illuminated from behind by a broadband quartz-halogen source through an aperture ($\sim 200 \mu\text{m}$ diameter) mounted just behind the polymer multilayer. A $50\text{-}\mu\text{m}$ diameter optical fiber is mounted on a two-axis translation stage that enables the fiber entrance face to be hand-aligned with the symmetry axis of the microlens. The microlens/aperture combination is mounted on an electrically-actuated stage (on a kinematic mount) in order to enable more careful alignment between the microlens and the optical fiber placed above it. Proper alignment is confirmed by the shape of the spectral output from the fiber as a function of distance from the microlens.

A 60-second ramp applied to the actuating stage then withdraws the microlens longitudinally from contact with the collecting fiber to a distance of up to 1 mm. During this motion, every 0.2 seconds the spectrometer (an OceanOptics USB4000 UVVIS, 0.3 nm resolution) records the spectrum. Repeated fine adjustments to the alignment of the microlens with the fiber are made while scanning along the optical axis through the focal length until the signal from the fiber peaks as it passes axially through the focal point of the microlens. Once optimally aligned, about 300 spectra are recorded during each of multiple ramp cycles. These spectra are then reorganized by individual wavelength, yielding an intensity versus distance dataset (we call a

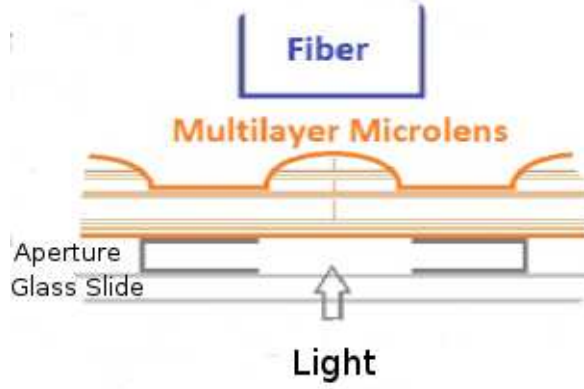


Fig. 2. Experimental setup for characterizing the spectral dispersion of the focusing properties of the microlenses.

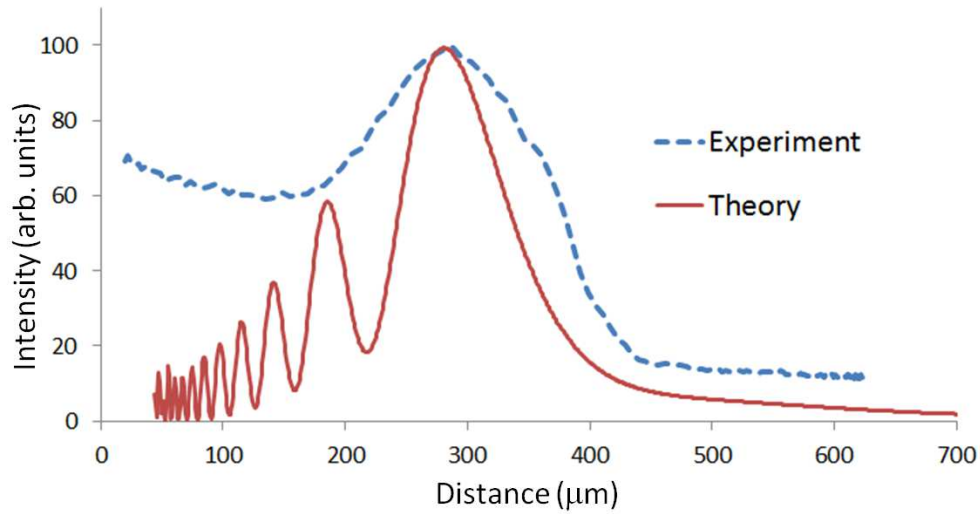


Fig. 3. The dashed blue trace is a typical multilayered co-extruded polymer microlens light curve of captured focused light. The wavelength for this light curve is 695 ± 5 nm and the focal length is ~ 300 microns. The solid red trace is an idealized light curve, from physical optics theory assuming monochromatic light (~ 695 nm) incident along the optical axis of the microlens whose dimensions are given in Section 4.

“light curve”) for each wavelength. A representative measured light curve (dashed blue trace) at a single wavelength is shown in Fig. 3 along with a typical theory curve (solid red trace). The location of the peak indicates the approximate focal length. The qualitative difference between the theory and experiment is primarily a consequence of the non-ideality of the lens and underlying multilayer structure as well as the geometry/acceptance of the fiber. The calibration of the distances in the actuating stage is accomplished with a micrometer, yielding a distance versus current calibration that is accurate to about 10% in determining the absolute focal length, but can fix the dispersion much more precisely.

In addition, the focused beam waist was measured under white light using a NIKON Eclipse

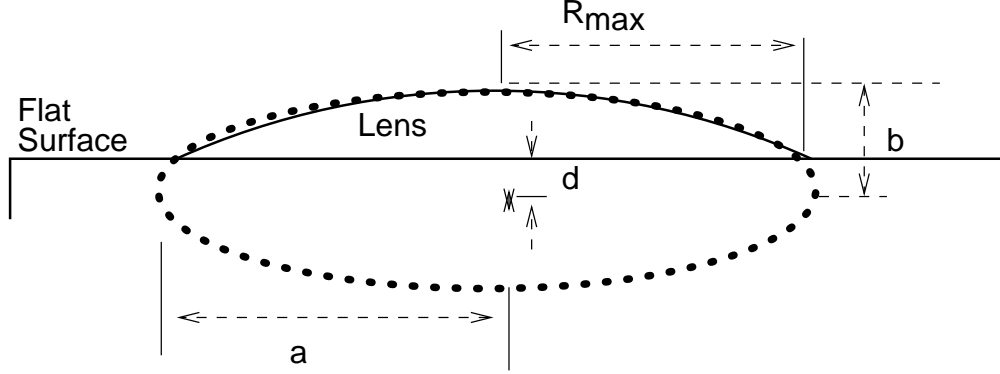


Fig. 4. Fixing the notation for a section of an oblate spheroid lens, showing ellipse semimajor (a) and semiminor (b) axes, the outer radius of the raised plano-convex lens shape R_{max} , and the depth of the center of the ellipse (d) below the plane of the surface supporting the lens.

M600 microscope (objective $100\times/0.9$) to determine the spot size on a 1.3 MPixels CMOS array and found to be $3.3 \pm 0.5 \mu m$, which is close to the approximate expected diffraction limit of the microlens ($\sim 3 \mu m$).

4. Theory and data analysis

To model the dispersive optical properties of the multilayered microlens, rather than full Finite-Difference Time-Domain (FDTD) simulation, we use Huygen's principle and add up the spherical waves coming from each section of the lens, while paying careful attention to the phase retardation at each surface due to the wave's propagation through the layered microlens [29].

Refer to Fig. 4 to fix the geometrical parameters we use to describe the lenses (*i.e.* as a section of an oblate spheroid formed by rotating an ellipse about its minor axis). In terms of these parameters, the radius of curvature at the center of the lens is $\tilde{R} \sim \frac{a^2}{b}$, and so, were it a monolithic lens of material index n , the thin lens approximate focal distance would be $f \sim \frac{a^2}{(b-d)(n-1)}$. Note that $R_{max} = a\sqrt{1 - \frac{d^2}{b^2}}$, in terms of which

$$\tilde{R} = \frac{R_{max}^2}{b-d} \frac{b}{b+d}, \quad (1)$$

an expression that smoothly interpolates between the elliptical and parabolic cross-section limits, in which $\tilde{R} = \frac{R_{max}^2}{2h}$ and $f = \frac{\tilde{R}}{(n-1)}$ and altitude $h = b - d$. By altitude we are referring to the maximum height of the microlens surface above the flat segment between lenses.

Because we are most interested in the far-field, we simply add up spherical waves

$$d\psi = z_h \frac{e^{ik_0 r}}{r} dA \quad (2)$$

emanating from each surface dA of the ring of height h making up the lens, with z_h being the magnitude and phase of the wave as it emerges from that surface (r is the distance to the point at which we are combining all of the waves) and where k_0 is the magnitude of the vacuum wavevector. Technically, the integration must include all of the illuminated surfaces, including the flat parts, but in the case of uniform illumination we can use linearity to reduce it to just an integral across the lens.

The final simplification we employ comes from the paraxial limit. In general, the computation of the far fields can be written as sums of elliptics or (in the case of azimuthal symmetry) Bessel functions [29]. In the paraxial limit, a distance $X \gg 2R_{max}$, but much closer to the optical axis than $X\lambda/R_{max}$, the integrations simplify and can be written in terms of exponentials. On axis, each annular section (of r_{outer} outer and r_{inner} inner radius) at height h of the lens contributes to the wave sum an amplitude S_h given by

$$S_h = \frac{1}{X-h} \int_{r_{inner}}^{r_{outer}} r dr e^{i \frac{k_0 r^2}{2(X-h)}} (z_h e^{-ik_0 h} - 1), \quad (3)$$

where we have removed an irrelevant overall phase factor and have taken the difference between the amplitude and phase of the wave emerging from that height of material and that amplitude (unity) and phase ($e^{ik_0 h}$) that would have emerged from there *were there no lens there at all*.

Thus, for each annulus that makes up the lens, the contribution it makes to the on-axis far field (at position X) is

$$S_h = \mathcal{A} (z_h e^{-ik_0 h} - 1) [e^{i \frac{k_0 r_{outer}^2}{2(X-h)}} - e^{i \frac{k_0 r_{inner}^2}{2(X-h)}}], \quad (4)$$

where \mathcal{A} is some overall amplitude which scales with the input wave's amplitude and is the same for all annuli. Thus, the total wave amplitude S at a distance X from the lens but on axis is $S_{tot} = \sum_h S_h$. These expressions are complicated to evaluate for layered materials because z_h is an intricate function of h . For the well known case of a monolithic plano-convex lens, however, one can compute this sum analytically, recovering in the thin lens limit the familiar results of a Gaussian beam focal distance and waist.

To find z_h rigorously would require that one solve Maxwell's equations for the wave field in this layered geometry, or obtain it numerically via a finite element (for example, via FDTD) simulation. More intuitively, we adopt an approximate z_h , namely, we estimate z_h from the transmission amplitude for this normal incident wave to exit a stack of these multilayers that would have terminated at h . Physically this approximation includes only internal reflections in the full multilayer from the layers below height h , discounting reflections from the rest of the lens that contribute to the wave emanating from the annulus at height h .

The modulus square of the resulting S_{tot} as a function of X , the distance along the axis, typically leads to a light curve as in Fig. 3 (red solid trace) as compared with the measured light curve (blue dashed trace) for one of the layered microlenses. Note that the theory trace can be thought of as plot of the intensity (essentially, $|S_{tot}|^2$) on the lens axis versus distance (X) from the lens. We identify the maximum of this light curve as the focal length of the lens at this wavelength, and note that at large X , ignoring background light, the intensity falls off universally as $1/X^2$, as expected. In experiment, the interference fringes at intermediate X are generally smeared out due to spectral and spatial averaging.

Equating the location of the on-axis light curve maximum with the focal length, theory indicates that these lenses will typically have large, designable chromatic aberration. The solid green trace of Fig. 5 is a typical theoretical curve depicting $\delta f/f$, the fractional variation in the focal distance with wavelength, for a microlens with (compare Fig. 1(d) and Fig. 4) $d = 0.27$ microns, $a = 34$ microns, $b = 2.5$ microns built from a 32-layer multilayer lens blank having a reflection band stretching from 430 nm to 510 nm (as shown in transmission curve (dashed red) of Fig. 5). In essentially all cases, the theory model predicts focal lengths for these multilayered convex lenses that are shorter on the red (long wavelength) side of the band edge than the blue side, opposite to the usual expectations of chromatic aberration in a solid lens of a material with normal dispersion. As described below, this shape can be understood in terms of the effects of the transmission band structure created by the multilayering (again, for example

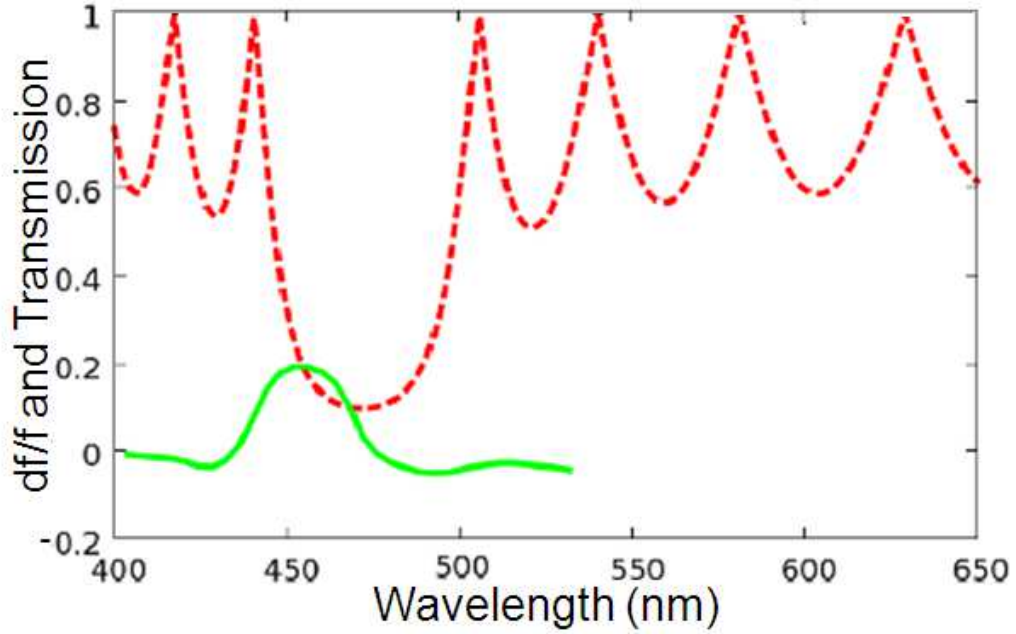


Fig. 5. Multilayered microlens computed focal length dispersion using physical optics theory described in the text. The dashed red trace is the computed transmission band of the 32 perfectly uniform layer model (layer indexes of 1.58 and 1.44) showing a reflection band between 440 and 510 nm. In solid green is the computed fractional change in the focal length (df/f) of the multilayer structure showing a profound change around the reflection band edge.

as shown by the dashed red trace in Fig. 5). FDTD calculations for a plano-convex shape in Fig. 4 reveal that there is little noticeable change in the focus spot size across the reflection band, and, further, that the spot size corresponds closely to that of a monolithic lens. As an additional check we note that these more detailed numerical calculations show the same qualitative changes in the focal length as the wavelength varies across the reflection band as are seen in the simple semi-analytical model described above (Fig. 5) and as noted in experiment (Fig. 6).

5. Discussion

Figure 6 shows the measured transmission band and focal length variations of a single multilayered PS/PMMA polymer microlens of radius $\sim 28 \pm 4$ microns and height about $2.5 \pm .5$ microns. These measurements were made using AFM and the stated variations are due to process variations across the microlens array, not dimensional uncertainty in the measurements of a single lens. In that figure, the shallow reflection band stretching from 430 nm to 510 nm is easily discernable, as is the pronounced chromatic aberration in the focal distance of nearly 30 percent in a span of 25 nm. The measured average focal length was $\sim 300 \pm 20$ microns. Note that this is broadly consistent with expectations for a thin lens of material whose index is the average of the constituent polymer indices. The scatter in the focal lengths at each wavelength is technical noise associated with the jitter in the numerics associated with determining the maxima of each of the experimental light curves. Fitting the portion of that df/f versus λ curve away from the reflection band reveals the nominal changes in the focal length expected

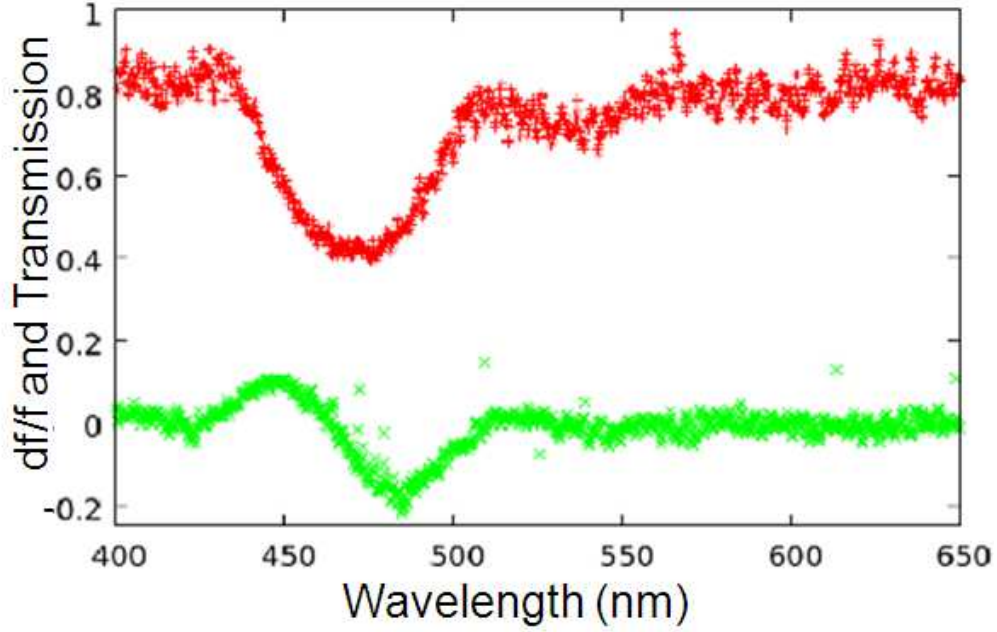


Fig. 6. Measured layered microlens focal length dispersion (green ‘x’) using the fiber transport system described in the text and the layered material’s transmission spectrum (red ‘+’). The lens material has an easily discernable shallow reflection band from about 430 nm to 510 nm. The len’s focal length dispersion is pronounced across the band, amounting to nearly 25% changes in the $\sim 300 \mu\text{m}$ focal length.

from dispersion in these bulk materials, albeit reduced somewhat from the diffractive effects from the smallness of the lens itself. For comparison, the expected (fractional, normal) chromatic aberration in a focal distance of a lens made from a (unlayered) PS/PMMA blend in this wavelength range (500-1000 microns) is expected to be roughly 2% per octave, and a linear fit in this wavelength range of our data is consistent with roughly 1% per octave. This and the regular shape of the light curves away from the material’s reflection band (Fig. 3, blue dashed trace) further support the identification of this feature with a microlens of the shape imaged in Fig. 1.

The qualitative similarity between the experimentally measured focal length dispersion and that derived from the simple physical optics model can be readily understood in terms of where the light lingers as it flows through the material. Recall that for these binary multilayers, at the short wavelength edge of an optical band, the light’s electric fields are primarily in the low index material whereas, at the long wavelength band edge, they reside primarily in the high index material. Thus, because the effective index that the light fields respond to at the long wavelength side is closer to the lower index material’s, the focal length is larger, and vice versa. This simple narrative also leads to an estimate for the upper bound of the focal length variation; because the focal length is inversely proportional to $(n - 1)$, we expect a change across the band of $(n_{\text{high}} - n_{\text{low}})/(\bar{n} - 1) \sim 20\%$ (for a PS/PMMA layered film), akin to that measured. Further discussion of errors due to layer thickness variations and imperfections due to the photolithography etching process are beyond the scope of this demonstration [12, 31].

6. Conclusions and future work

We have created layered microlenses with designed chromatic aberration in the visible. A simple physical optics model for the transmission and focusing of the light traversing these microlenses shows how the ‘structural’ dispersion caused by the layering leads to this designed chromatism. Even though such chromatism occurs in the multilayer material’s reflection band, the number and properties of the layers can be designed and tuned to still allow a substantial percentage of the light through, while preserving the chromatism. We have studied these microlens structures primarily as a test of our overall understanding of the role of designed dispersion in layered microlenses, with potential applications in multi-spectral imaging, and dispersion correction and control.

Our work here builds upon earlier work demonstrating the benefits of a roll-to-roll co-extrusion process for multilayering to create Bragg reflectors [27,30] and distributed feedback lasers [23,25,32]. We emphasize that the simple multilayer stack design used is not necessarily ideal for applications and we did not investigate these materials for imaging applications, but we have used this system to better understand the feasibility of the novel combination of an axially-cut, lens-like pattern in a multilayer polymer.

Beyond the polymer combinations used here, this work connects the designed optical dispersion in metamaterials in the visible with the properties of a lens. Depending upon the parameters of the etching process, the resulting shape may be smoothly varying through the layers or can be etched in a terraced layer-cake structure by taking advantage of different etching speeds in the different polymers. In an effort to understand more fully the connection between multilayer design and the chromatic properties of the lens, we are currently trying to make multilayered lenses with higher numerical aperture as well as exploring novel polymer multilayer optical materials such as those with designed phase-slip defects formerly used in laser and magneto-optics studies [30,32]. Optical transport in these phase-slip defect multilayers is well understood and will make lenses ideal for more rigorous testing of our model. Because we can also create multi-band, gradient, and ‘chirped’ structures as described in [33], we are exploring the utility of lenses made from materials with more intricate customizable dispersion. For future development, we note that a grayscale mask can be used with multilayers to create a circular grating structure that has been shown to be an attractive structure for confining modes in a surface-emitting photonic bandgap laser [34]. Combining the surface structuring with the multilayer Bragg structure is one approach to a more easily fabricated three-dimensional photonic crystal.

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